# Nuclear Criticality Safety Engineer Training Module 8

# Hand Calculation Methods - Part I Buckling Conversion and Surface Density Methods

### LESSON OBJECTIVE

This module presents two traditional hand calculation methods for estimating the critical size or subcriticality of a system, or establishing the limiting conditions for either single fissile units or arrays of units.

# **REFERENCES**

Many textbooks provide derivations of buckling parameters. The surface density model is described in standard criticality safety references and many journal articles. A few of the readily available references are listed below.

- 1) J. J. Duderstadt and L. J. Hamilton, <u>Nuclear Reactor Analysis</u>, John Wiley and Sons, Inc., New York (1976).
- 2) Ronald Allen Knief, <u>Nuclear Criticality Safety Theory and Practice</u>, American Nuclear Society, La Grange Park, Illinois (1991).
- 3) J. T. Thomas, "Nuclear Safety Guide TID-7016, Revision 2," NUREG/CR-0095 ORNL/NUREG/CSD-6, Oak Ridge National Laboratory (June 1978).
- 4) H. C. Paxton, J. T. Thomas, Dixon Callihan and E. B. Johnson, "Critical Dimensions of Systems Containing U<sup>235</sup>, Pu<sup>239</sup> and U<sup>233</sup>," Los Alamos Scientific Laboratory and Oak Ridge National Laboratory Report TID-7028 (June 1964).
- 5) H. C. Paxton and N. L. Pruvost, "Critical Dimensions of Systems Containing U<sup>235</sup>, Pu<sup>239</sup> and U<sup>233</sup>, 1986 Revision," Los Alamos National Laboratory Report LA-10860-MS (July 1987).

### **BACKGROUND**

Before the development of high-speed, large-memory computers, hand calculations were widely used to establish safe limits for fissile material operations. Today these methods might seem obsolete, but they are still useful in providing a starting point for more elaborate calculations or for providing a "sanity check" on the results of complicated computations. A preliminary review of a fissile material system using one or more of these methods can often provide insight into the

problem that may not be apparent from simply looking at a computer printout. These methods also have a historical importance and provide a glimpse of the early days of criticality safety calculations.

Part I of Hand Calculation Methods discusses buckling conversions and the surface density model. Part II will cover the density analog and limiting surface density methods.

# **BUCKLING CONVERSIONS**

The buckling conversion method is based on solutions to the diffusion equation that relate the spatial neutron flux distribution in a neutron-multiplying medium to a parameter called the geometric buckling,  $B_g^2$ . One of the earliest uses of this method was to relate the critical dimensions of spheres, that had been measured experimentally, to those of cylinders of various shapes (Ref. 4). Derivation of the buckling equations can be found in many nuclear engineering text books (e.g., Ref. 1) and will only be summarized here. A concise derivation can also be found in NCSET Module 6.

#### THE DIFFUSION EQUATION

The general diffusion equation in a multiplying medium is

$$D\nabla^{2}\phi(\bar{\mathbf{r}},t) - \Sigma_{a}\phi(\bar{\mathbf{r}},t) + \nu\Sigma_{f}\phi(\bar{\mathbf{r}},t) = \frac{1}{\nu}\frac{\partial\phi(\bar{\mathbf{r}},t)}{\partial t}.$$
 (1)

 $\phi(\bar{r},t)$  is the neutron flux and the diffusion coefficient, D, in this approximation is

$$D = \frac{\sum_{s}}{3\sum_{t}^{2}}$$
 (2)

with the subscripts s and t indicating scattering and total cross sections, respectively.

This partial differential equation can be solved by using the separation of variables method to look for a solution of the form

$$\varphi(x,t) = \Psi(x)T(t).$$

Considering only one dimension, the spatial part of the diffusion equation is

$$D\frac{d^{2}\Psi}{dx^{2}} + \left(\nu\Sigma_{f} - \Sigma_{a}\right)\Psi(x) = -\frac{\lambda}{\nu}\Psi(x)$$
(3)

where  $\lambda$  is a constant to be determined. As in Module 6, consider a homogenous problem similar to the spatial diffusion equation,

$$\frac{\mathrm{d}^2\Psi}{\mathrm{d}x^2} + \mathrm{B}^2\Psi(x) = 0 \tag{4}$$

with the boundary conditions

$$\Psi\left(\frac{a'}{2}\right) = 0$$
 and  $\Psi\left(-\frac{a'}{2}\right) = 0$ .

B is an arbitrary parameter and a' represents an extrapolated dimension of the system (e.g., x + d, with d being the extrapolation distance). This problem actually has a series of solutions, but the fundamental mode is

$$\Psi(\mathbf{x}) = \mathbf{A}_1 \cos\left(\frac{\pi \mathbf{x}}{\mathbf{a}'}\right). \tag{5}$$

If we identify this eigenvalue problem with the space dependent diffusion equation, then

$$\mathbf{B}_{1}^{2} = \frac{1}{\mathbf{D}} \left( \frac{\lambda_{1}}{\mathbf{v}} + \mathbf{v} \Sigma_{f} - \Sigma_{a} \right)$$

or

$$\lambda_1 \equiv v\Sigma_a + vDB_1^2 - vv\Sigma_f. \tag{6}$$

It is customary to refer to the value of B<sub>1</sub><sup>2</sup> for the fundamental mode as the *geometric buckling* 

$$\mathbf{B}_1^2 = \left(\frac{\pi}{\mathbf{a}'}\right)^2 \equiv \mathbf{B}_{\mathrm{g}}^2 ,$$

where the factor  $\pi/a'$  is characteristic of cartesian coordinates.

By definition, the state of criticality implies a time-independent flux distribution, which in turn requires that the fundamental eigenvalue vanish.

$$\lambda_1 = 0 = v(\Sigma_a - v\Sigma_f) + vDB_1^2$$

The solution becomes

$$\phi(x,t) \rightarrow A_1 \cos B_1 x \neq \text{ function of time}$$

with the criticality condition

$$B_1^2 = B_g^2 = \frac{v\Sigma_f - \Sigma_a}{D} = B_m^2 . ag{7}$$

# MATERIAL AND GEOMETRIC BUCKLING

That is, at critical, the material buckling,  $B_m^2$ , which depends on the material properties (cross sections and atom densities) equals the geometric buckling  $B_g^2$ , which depends on the geometry (e.g., slab width, a', in one-dimension cartesian coordinates). Thus, if one knows the critical geometry for a sphere of a given material, the material buckling can be calculated and that can be used to determine the critical geometry for other configurations of the same material.

Expressions for the geometric buckling of various geometries are given in many text books and references. Some of the common ones are listed below (in each case,  $\lambda$  is the extrapolation distance) and others can be found for hemispheres, semi-infinite cylinders, cylinders with polygonal cross sections and more.

Infinite slab of width a: 
$$B_g^2 = \frac{\pi^2}{(a+2\lambda)^2}$$
 (8)

Cuboid with sides a, b, and c: 
$$B_{g} = \frac{\pi^{2}}{(a+2\lambda)^{2}} + \frac{\pi^{2}}{(b+2\lambda)^{2}} + \frac{\pi^{2}}{(c+2\lambda)^{2}}$$
 (9)

Sphere of radius r: 
$$B_g^2 = \frac{\pi^2}{(r+\lambda)^2}$$
 (10)

Finite cylinder with radius r and height h: 
$$B_g^2 = \frac{\pi^2}{(h+2\lambda)^2} + \frac{(2.405)^2}{(r+\lambda)^2}$$
 (11)

The extrapolation distance must be included because diffusion theory over-predicts critical dimensions. The extrapolation distance (or extrapolation distance plus reflector saving for reflected systems) must be subtracted from the calculated dimension or added to the physical dimension in the expressions for  $B_{\rm g}^{\ 2}$ .

Figures 8-1 and 8-2 show typical curves of extrapolation distances for sphere and cylinders of several materials. These curves are taken from TID-7028 (Ref. 3). Each curve has points for spheres plus curves for cylinders with varying height-to-diameter ratios. Note the values for spheres lie at H/D=1.

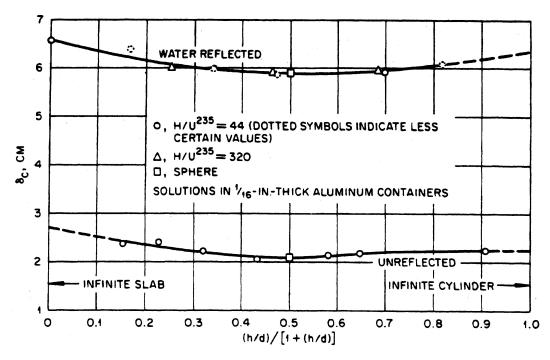


Figure 8-1. Effective extrapolation distances for cylinders of U(93.2)O<sub>2</sub>F<sub>2</sub> solutions. Cylinder height and diameter are h and d, respectively. (From TID-7028, Fig. 3)

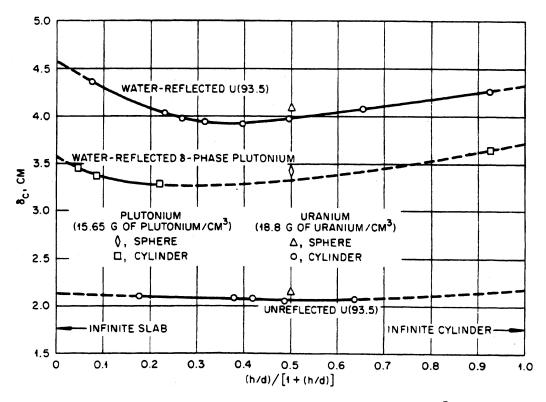


Figure 8-2. Effective extrapolation distances for cylinders of U(93.5) and  $\delta$ -phase plutonium metal. Cylinder height and diameter are h and d, respectively. (From TID-7028, Fig. 4)

#### A BUCKLING EXAMPLE

Consider an example of using buckling conversion to calculate the relation between the critical mass of a water-reflected sphere of enriched uranium metal (93.5 wt-%  $^{235}$ U at a density of 18.8 g(U)/cm³) and the critical masses of water-reflected cylinders containing the same material. Since the buckling of the cylinder is a function of both height and diameter, this is a parametric problem with a series of solutions. Figure 42 of Ref. 5 gives the critical mass of a water-reflected sphere of this material as approximately 22.2 kg. Based on the given density, the radius of this sphere is 6.56 cm. Using the top curve in Fig. 8-2,  $\delta$  = 4.1 cm for the sphere. Combining these values, Eqn. 10 gives  $B_g^2$  = 0.0868 cm².

Next, pick a height-to-diameter ratio, for example, 5. From Fig. 8-2 the extrapolation distance for this cylinder is about 4.24 cm (tip: use a ruler to interpolate between grid lines). Knowing the buckling and H/D, Eqn. 11 can be solved for either r or h. This is most easily done by using a program with an equation solver, such as Excel. Doing this, the radius is calculated to be 4.12 cm, giving a critical mass of 41.2 kg. Note that this long, thin cylinder has a critical mass approximately twice that of the sphere. This result is in good agreement with Fig. 5 of Ref. 5.

If this procedure is repeated for several values of H/D, a curve similar to the one below can be generated. Note that the minimum critical mass of the cylinder has a height-to-diameter ratio of unity. Also note that the critical mass of the sphere is lower than that for the optimum cylinder, reflecting the fact that the sphere is still a more reactive geometry.

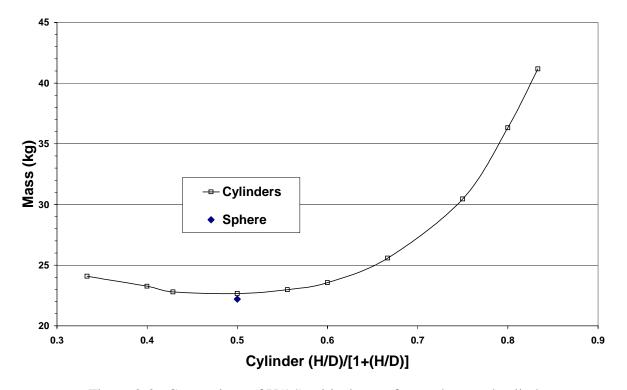


Figure 8-3. Comparison of U(94) critical mass for a sphere and cylinders.

#### SURFACE DENSITY METHOD

The basis of the surface density method is a comparison of the fissile material density of an array as projected onto a plane with the density of a water-reflected critical slab of the same material. This method is useful for determining safe parameters for arrays including the fissile mass per array unit, spacing of the units or the maximum safe number of units that can be stacked. Similar methods such as the density analog and limiting surface density models will be covered in the second NCSET module on hand calculations.

The surface density method is semi-empirical, being based on a series of experiments plus calculations [3]. Many formulations of this method exist with varying underlying assumptions. Features of multiple methods must not be combined without adequate validation of the resulting method. One well-accepted formulation of the surface density method, on which the following discussion is based, is given in Ref. 3. The data required to use this method are the critical density of a water-reflected infinite slab and the critical mass of an unreflected sphere of the material in question.

Given that the mass of an array of fissile material is projected onto a plane surface to form the equivalent of a slab of that material, the allowed (i.e., subcritical) projected surface density (in g/cm²) is given by the expression

$$\sigma = 0.54\sigma_0 (1 - 1.37f) \tag{12}$$

where  $\sigma_0$  is the surface density of the critical water-reflected infinite slab (in g/cm²) and f is the fraction critical of a unit in the array, i.e., the ratio of the mass of the unit to the critical mass of an unreflected sphere of the same material. (Note the use of the water-reflected slab density, but the bare spherical critical mass.) Since the surface density cannot be negative, the fraction critical is limited to a value of about 0.73 in this formulation of the method.

If the array of units, each with fissile mass m (in g), is considered to consist of cubic volumes in a regular array, the projected surface density is

$$\sigma = \frac{nm}{d^2} \tag{13}$$

where n is the number of units stacked perpendicular to the projection plane and d is the length of the sides of the cubical volume associated with each unit. Combining these two expressions for  $\sigma$ , the safe dimension of the unit volume (in cm) is given by

$$d = 1.36 \left[ \frac{n \, m}{\sigma_0 \left( 1 - 1.37 f \right)} \right]^{1/2}. \tag{14}$$

In the absence of critical data, subcritical values from the figures in standard references, such as Ref. 3, may be used to give conservative results. According to Ref. 3, these equations are "applicable to infinite planar arrays reflected by water at least 155 mm thick or its nuclear equivalent." The reflector can be "no closer to units in the array than the boundaries of the cells associated with the units." This method can be used for arrays of bare metal, dry materials or solutions. The key is to use the data corresponding to the material or mixture in the array elements.

## A SURFACE DENSITY EXAMPLE

The surface density method can often provide a quick, conservative answer about the safety of an array. Consider a waste management organization that wants to store waste materials that contain 93 wt-% enriched UO<sub>2</sub> mixed with various plastics, papers, etc. Their plan is to use cubical steel bins, 4-ft on edge, and stack them three-high. The proposed <sup>235</sup>U mass limit is 325 g per bin. Is this a safe configuration?

The data needed to answer the question are the mass of a bare critical sphere of  $UO_2$  and the critical thickness of a water-reflected slab of the same material. Since no restrictions were put on the amount of moderating materials that could be mixed with the uranium, the worst case or optimal moderation must be assumed. From Fig. 2.1 of Ref. 3 (or from Module 5), the subcritical limit for a spherical mass of  $^{235}U$  in an aqueous  $UO_2$  solution or as a metal-water mixture is about 620 g, with a 25-mm thick reflector at a uranium concentration of about 0.044 kg(U)/L. This provides a conservative value for an unreflected critical sphere. From Fig. 2.4 of Ref. 3, the subcritical thickness limit for a reflected infinite slab of  $^{235}UO_2$  at the same uranium concentration is about 96 mm.

Using the spherical mass and the proposed mass limit, the fraction critical is f=0.524. The allowed surface density is the product of the slab thickness and the uranium density,  $\sigma_0=0.422$  g/cm². Putting these values into Eqn. 12, the limiting array surface density is 0.064 g/cm². Next, solve Eqn. 13 for n to get n=2.94. So the answer to the question about a stacking these bins three-high is that it is probably safe, but might require a little less conservative analysis. Subcritical limits were used for both the sphere and the slab, the value for the sphere was actually for a thin reflector, and no credit was taken for the enrichment of the material. Reducing the conservatism in these factors would probably give a value of n greater than three.

### **SUMMARY**

This module has presented two simple hand calculation methods, buckling conversion and surface density. Although largely displaced by the availability of today's fast computers, both methods are still used for quick, generally conservative estimates of the criticality safety of both single fissile units and arrays.

# **PROBLEMS**

- 1. A glove box used to machine samples of uranium (94 wt-% <sup>235</sup>U) has a small rectangular sump, 8 in. x 8 in. x 8 in. deep, in one corner to collect water from wet grinding operations. Is this sump geometrically safe? Assume the extrapolation distance for a cuboid is the same as that for a cylinder.
- 2. Table 29 of LA-10860-MS gives the critical mass of a bare sphere of U(94) with a density of  $18.74 \text{ g/cm}^3$  as 49.12 kg. Calculate the radius and geometric buckling of the critical sphere. Calculate the mass and radius of a cylinder of U(94) with H/D = 1 having the same buckling.
- 3. A facility would like to store units of 5 kg of  $U(70)O_2$  with H:U = 12 in stacks two-high. Calculate the limiting surface density and the recommended array spacing.

#### PROBLEM SOLUTIONS

1. From Fig. 7 of LA-10860-MS, the extrapolation distance for unreflected U(94) cylinders is nearly constant at 2 cm. Using the given assumption that the extrapolation distance is the same for a cuboid, the buckling of the sump is (from Eqn. 9)

$$B_{\text{sump}}^2 = 3 * \frac{\pi^2}{(20.32 + 2 * 2)^2} \text{cm}^{-2}$$
$$= 0.050 \text{ cm}^{-2}$$

From Fig. 6 of LA-10860-MS, the ratio of the extrapolation distance for a sphere to that for a cylinder is nearly one. From Eqn. 10, the radius of an equivalent sphere is then

$$B_{\text{sphere}}^2 = \frac{\pi^2}{(r+\lambda)^2} = 0.050 \,\text{cm}^{-2}$$

Solving this equation, r = 12.05 cm and V = 11.6 L. Figure 2.2 of TID-7016 Rev. 2 shows that the minimum critical volume for  $^{235}$ U can be much less than 11.6 L, so this sump is not geometrically safe.

2. Since the volume of the sphere is simply the mass divided by the density, V = 2.62 L. The radius can then be calculated as 8.553 cm. Figure 7 of LA-10860-MS shows that the effective extrapolation distance for a cylinder with H/D = 1 is about 2 cm for 94% enriched uranium. Figure 6 of the same reference shows that the ratio of the spherical extrapolation distance to the cylindrical extrapolation distance is nearly 1 for cylinders with H/D = 1. Since the ratio of the spherical to cylindrical extrapolations distances is 1, and the spherical extrapolation distance is given as 2 cm, Equation 10 gives

$$B_g^2 = \frac{\pi^2}{(8.553 + 2)^2} = 0.0886 \,\text{cm}^{-2}$$

With this value of the buckling, and assuming that the axial and radial extrapolation distances for the cylinder are the same, Equation 11 can be used to find the cylinder radius by inserting the buckling calculated above with  $\lambda=2$  cm, then solving for r=7.65 cm. Since H/D = 1, the height of the cylinder is 15.3 cm, the volume is 2.81 L and the mass is 52.7 kg, slightly larger than the spherical critical mass.

3. This is the sample problem given in TID-7016 Rev. 2. Using the 25-mm reflected curve from Fig. 2.1 of that reference, a conservative value for the critical mass of an unreflected sphere is approximately 14 kg (U). (Note that this is for <sup>235</sup>U. According to Fig. 3.4 of TID-7016 this value could be increased by a factor of 1.5 for 70% enriched material.) From Fig. 2.4 of TID-7016 the reflected critical slab thickness is about 34 mm (also conservative since this is a

subcritical limit). At H:U=12, the density of uranium in the solution is about  $2.1~g(U)/cm^3$ . The surface density of the critical slab is then

$$\sigma_0 = 2.1 g(U) / cm^3 * 3.4 cm$$
  
= 7.14 g(U) / cm<sup>2</sup>

The fraction critical, not accounting for the enrichment, is f = 5/14 = 0.357. Then using Eqn. 12, the allowed surface density is

$$\sigma = 0.54 * 7.14 * (1 - 1.37 * 0.357)$$
$$= 1.97 \text{ g(U)}/\text{cm}^2$$

Eqn. 14 then gives, for the two-tier array,

$$d = 71.2 \text{ cm}$$
.

If the mass factor for the U(70) sphere is used, the fraction critical becomes 0.238, the surface density becomes  $2.6 \text{ g(U)/cm}^2$  and the allowed spacing is 62.0 cm.